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MON-H-258

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Contract No. W-35-058, eng. 71

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CLASSIFICATION CANCELLED
DATE JUN 17 1957 RLJ
For The Atomic Energy Commission
H. F. Carroll
Chief, Declassification Branch

RADIOACTIVE FISSION PRODUCT CONTAMINATION IN THE MUD OF WHITE OAK DRAINAGE SYSTEM

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Date Received: 3-20-47 Date Issued: 3/20/47

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J. S. Cheka and K. Z. Morgan

The White Oak Creek drainage basin is used for the disposal of radioactive wastes. The water is purified to a large extent by the precipitation and adsorption on clay and organic material of many of the radioactive isotopes. Deposition of activity has varied with changes in plant processes. During 1944, when Pu separation was the chief process, deposition ratio between the marsh at the upper end of the basin and the mouth of the White Oak Creek differed by a factor of about 10^4, the marsh having values up to 0.4 μc/gr. of surface mud. During 1945, when Ba separation was the chief process, values at the marsh dropped to less than half of the 1944 values, but the deposition ratio differed by a factor of less than 1000 between the above mentioned points. An assay of total curie content in the basin in April 1945 showed about 70 curies still being retained by the mud. This figure is 13% of the estimated total activity released into the basin between the beginning of operations and the time of the survey. Chemical analyses indicate that the distribution of isotopes also changed; Ba, Sr and Cs totals being less than 1% in 1944, and becoming about 50% late in 1945, Zr and Cb totals, meanwhile, dropping from about 75% in 1944 to about 2% late in 1945.

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Introduction

The purpose of this paper is to describe the radioactivity retained by mud in the White Oak drainage system of Clinton Laboratories. Another paper[1] describes the radioactivity of the discharged water.

Figure 6 indicates the principal components of the White Oak drainage system as (1) the settling pond, (2) upper dike, (3) marsh section, (4) intermediate pond, (5) lower dike, (6) White Oak Lake mud flats, (7) White Oak Lake, (8) marsh below White Oak Lake, and (9) the Clinch River. The various barriers to water flow in this system were provided with very little effort or cost since the entire White Oak drainage system is within the Oak Ridge restricted area. This system permits the removal of considerable fractions of most of the radioactive elements from the water by mud and plant life before the waste water enters the Clinch River. This system of marsh sections and mud flats is largely furnished by nature and has the advantage of requiring a minimum of upkeep effort. It is true that the long lived radioisotopes tend to build up concentrations in the mud in a manner that they can be partly washed down stream during flood season. However, measurements indicate that the increase in dilution water during a heavy rain usually more than compensates for the increase in the rate of discharge of radioactive materials at such a time. As a result, the curies per milliliter discharged into the Clinch River do not change very much, even with an increase in the rate of flow of White Oak Creek by a factor of perhaps 50 during flood periods. In the normal course of flow of White Oak Creek there is a dilution factor of about 8 in the creek between where the plant waste enters and where it empties into the Clinch River. The total normal dilution factor between the time waste water leaves the settling pond and mixes in Clinch River is about 3,000.

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1. Topography and Function of the Drainage Basin

The marsh formed by the two above mentioned dikes is approximately 2,000 feet long, and it takes about 4.5 hours for water to flow between these dikes by the most direct currents. The decontamination which began in the settling pond continues here; partly by a further settling of suspended precipitates, and partly by adsorption of fission-products to the alumino-silicates of the clay which forms most of the bed of the marsh. Adsorption takes place by ion exchange,[2] the heavier elements and those with higher charges having greater binding energies, and thus remaining fixed. There is also a possible flocculation of positively charged colloids due to interaction with negatively charged clay. Drs. Overstreet and Jacobson also made measurements[3] which indicated that less than 5% of the total activity from the W-6 waste tank escapes adsorption on Clinton clays. Tellurium is the only fission product which is not almost completely taken up by the clay. Studies made on plants indicated that growing plant roots also serve to remove fission products from the water and clay and fix them on the root surfaces. In the case of strontium, a large fraction is translocated in the stems and leaves.

The White Oak Dam backs up the creek about 2,000 ft. with the upper gate open, and about twice that distance with the upper gate closed. The lake thus formed has an area of about 17 acres with the upper gate open and about 32.5 acres with the upper gate closed. Passage of water along the main channel was measured by the use of dyes and it was found that it requires about 20 hours with the top gate open for water to flow from the settling pond to White Oak Dam. This time is, of course, subject to variation with rainfall. On the other hand, as the rainfall increases, the dilution increases and the water spreads out slowly over a widening area of mud flats and marsh sections.

Due to the numerous barriers along the White Oak system, there is little tendency toward streaming, and the active mud stirred up in the main creek channel spreads slowly and rather uniformly over an increasing impounding area.

Previous to October, 1944, a small pond, at point #5 on the map shown in Fig. 6, acted as a trap for entrained flakes of precipitates. The intermediate pond at the lower end of the marsh acts as a sort of trap for silt which may be washed from the rest of the marsh during heavy rains. It is 1.2 acres in area, 1 to 3 ft. in depth and has a series of ridges across the main channel, which act as baffles. There is a 4 to 8 in. deposit of silt throughout most of this pond.

2. Sampling and Counting

Routinely collected mud samples are obtained from the top 0.5 cm. of the creek bed, lake bottom and mud banks. Each sample is thoroughly mixed and a dab is uniformly spread on a petri dish and dried. After weighing, these are counted in a beta chamber, using a mica window counter. Corrections are made for background, scattering and geometry of the counter. These corrections are determined by comparison with a standard beta source. Correction is made for self-absorption of the mud, which will be discussed later. Counts per minute per gram are converted to microcuries per gram using the formula:

$$\mu c/gm = \frac{\text{counts/min/gm}}{\text{counter factor}} \times \frac{1}{60 \times 3.7 \times 10^4} \times \text{self absorption coefficient.}$$

3. Surveys and Findings

The first mud surveys on record are those of Overstreet and Jacobson[2] on 4/26/44 and 5/1/44. The high contamination of the creek bed shown by the first of these surveys motivated the discontinuance of the retention ponds and the construction of the settling pond described by Morgan and Western.[1] Discharge into the drainage system was stopped on 4/26/44 and was resumed on 7/3/44. Since that time, surveys have been made regularly of the mud and water activity.

Figures 1 and 2 show the distribution of mud activity throughout the White Oak drainage system for 1944 and 1945 respectively. As can be seen from the graphs, the sampling points were grouped according to the nature of location and distance from the settling pond. It was considered that the average of a similar group of samples was more representative of true values than the values from single locations. Figure 2 representing 1945 has more points shown than Figure 1 because the early surveys were not as complete. A comparison of the rainfall chart of Figure 3 with Figure 2 indicates that for 1944 the periods of heavier rains caused a greater amount of activity to reach the lower areas of the basin. The half-distance, or the distance downstream from the settling pond at which the activity drops to half its value, is about 1,200 feet in April and August when the rainfall is greater than 3 inches/month and about 800 ft. in July and October when rainfall is less than 3 inches/month. A comparison of these values with those of 1945 is not valid because of the frequent changes in constituents of the chemical wastes after discontinuance of the plutonium separation operations in the 205 building.

Figures 3 and 4 are summaries of mud data for 1944 and 1945. These graphs indicate how the radioactivity at a given location varies with time and with conditions at the time. Figure 3 covers grouped sampling points down to the upper end of White Oak Lake, and Figure 4 the points in lower White Oak Lake and below. Notations are made, showing the times at which major

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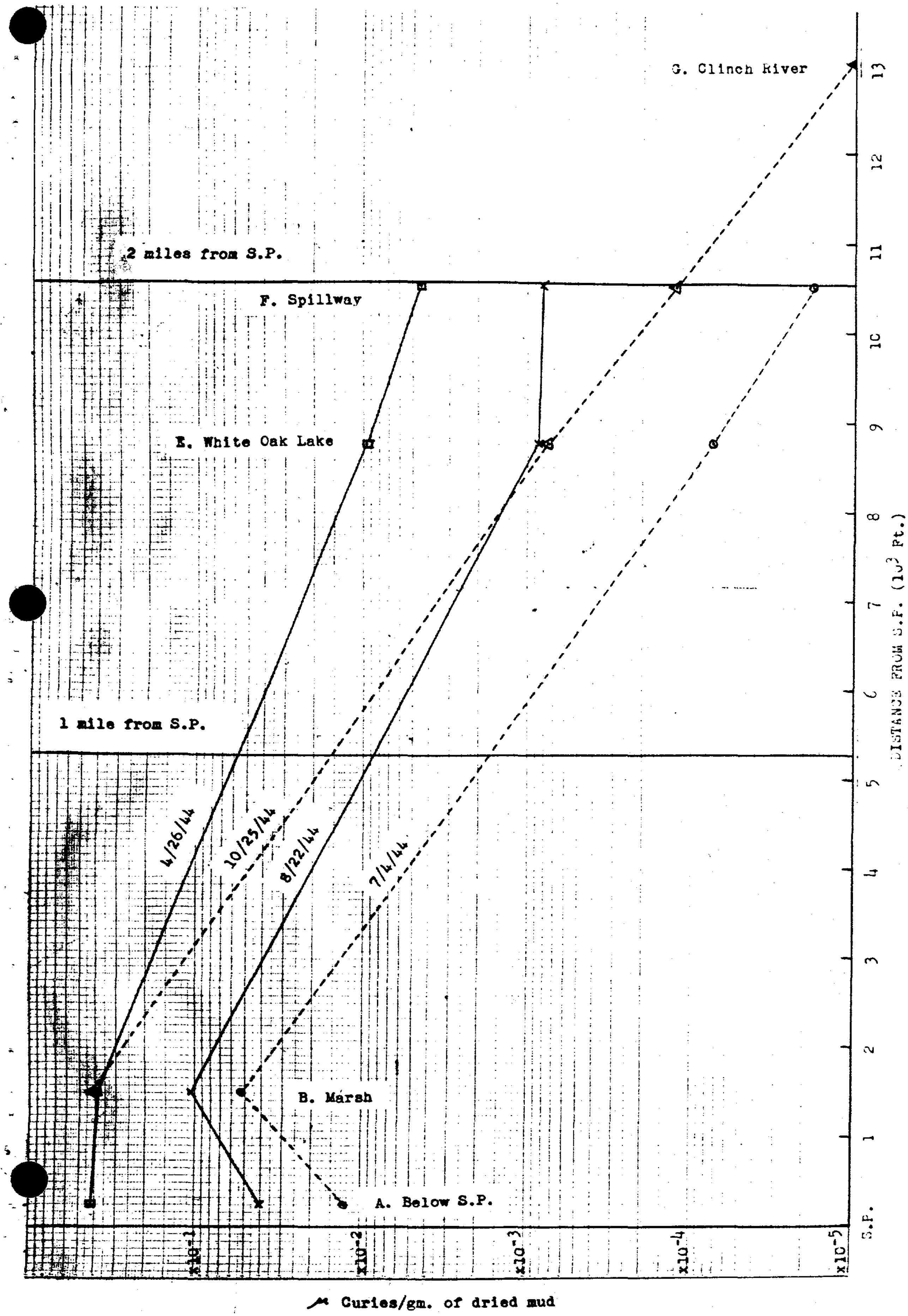

Fig. 1 MUD ACTIVITY DISTRIBUTION IN WHITE OAK CREEK --1944

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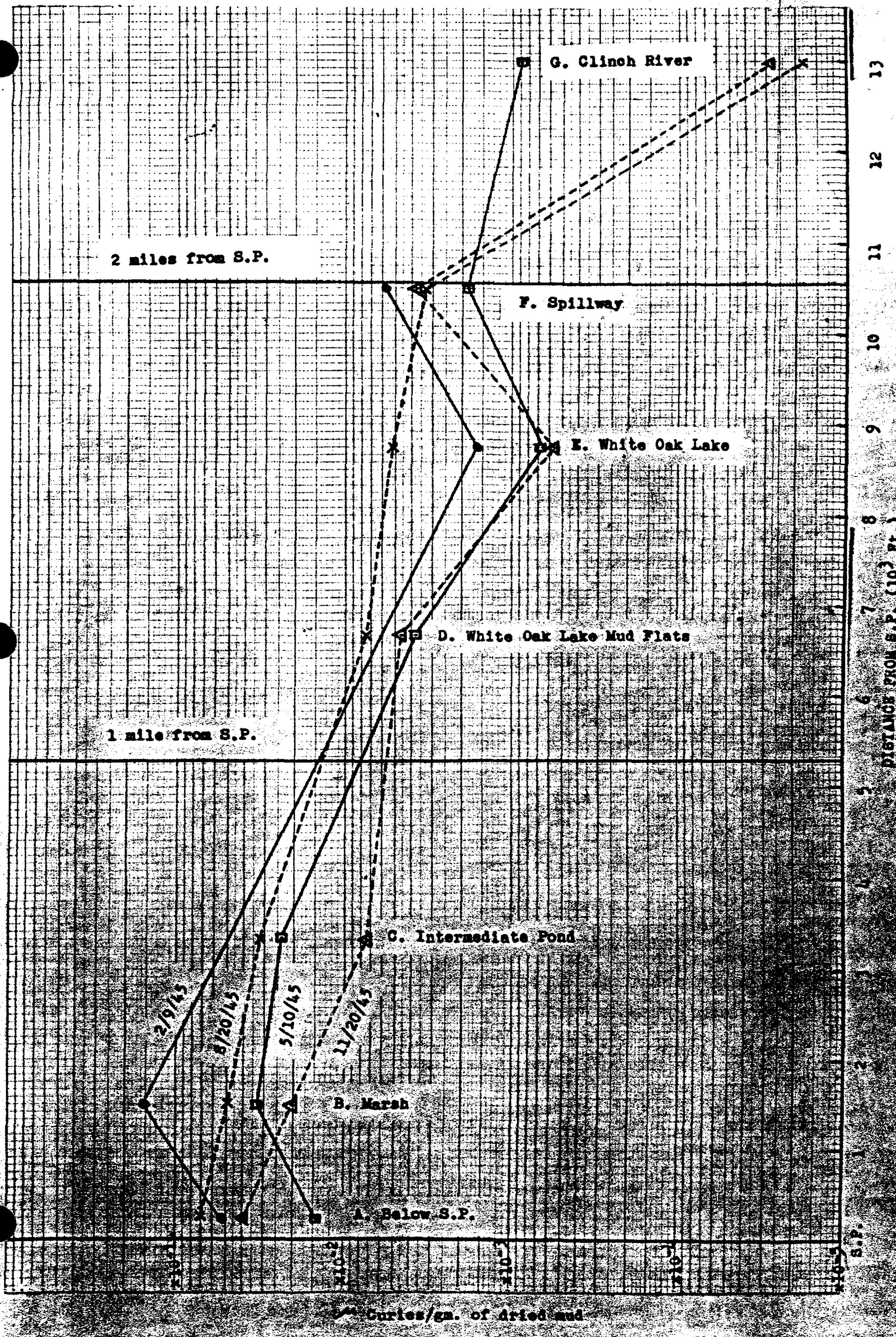

FIG. 2 MUD ACTIVITY DISTRIBUTION IN WHITE OAK CREEK — 1945

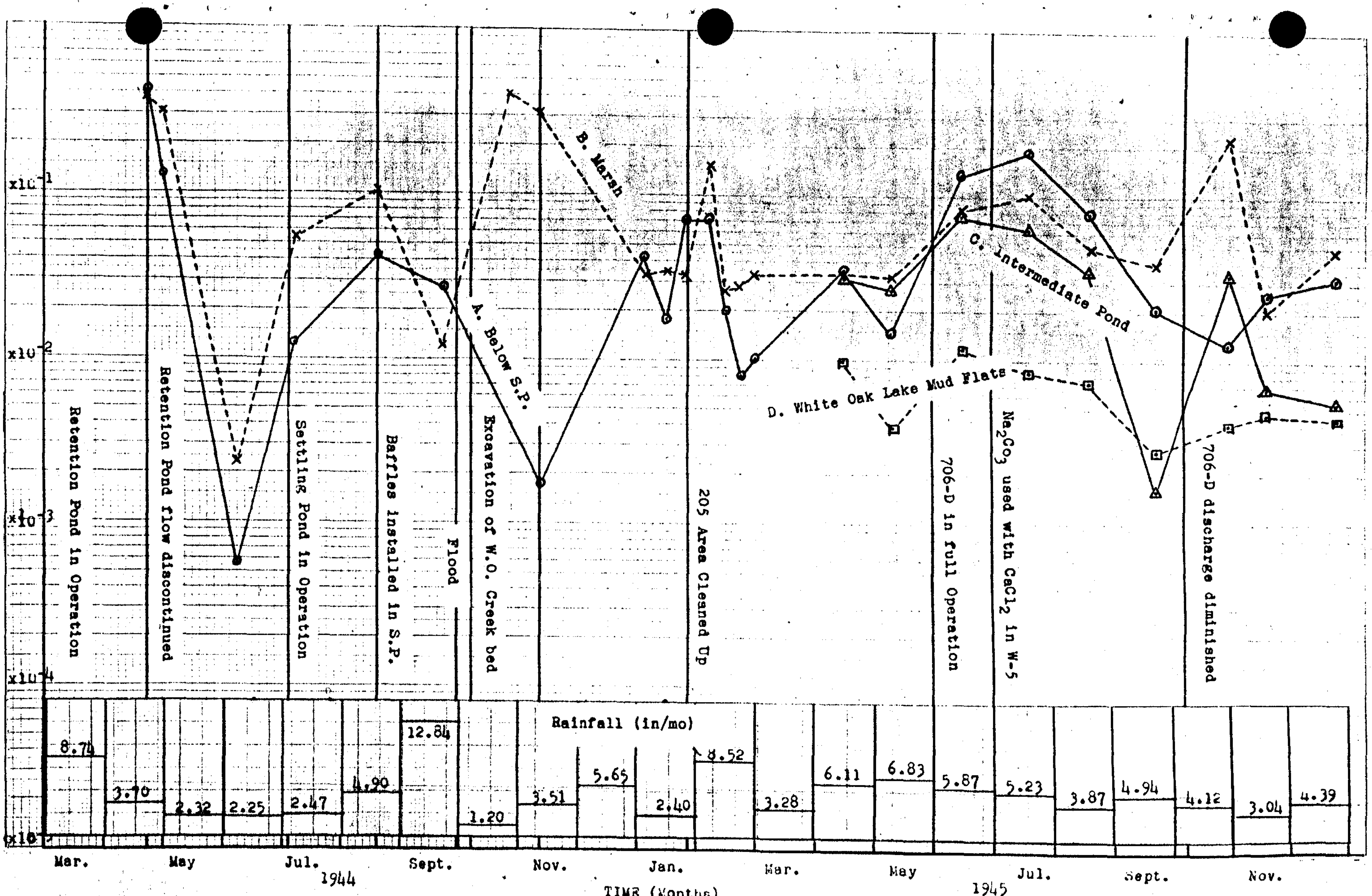

Fig. 3 SUMMARY OF WHITE OAK CREEK MUD DATA (above W.O. Lake) 1944 & '45

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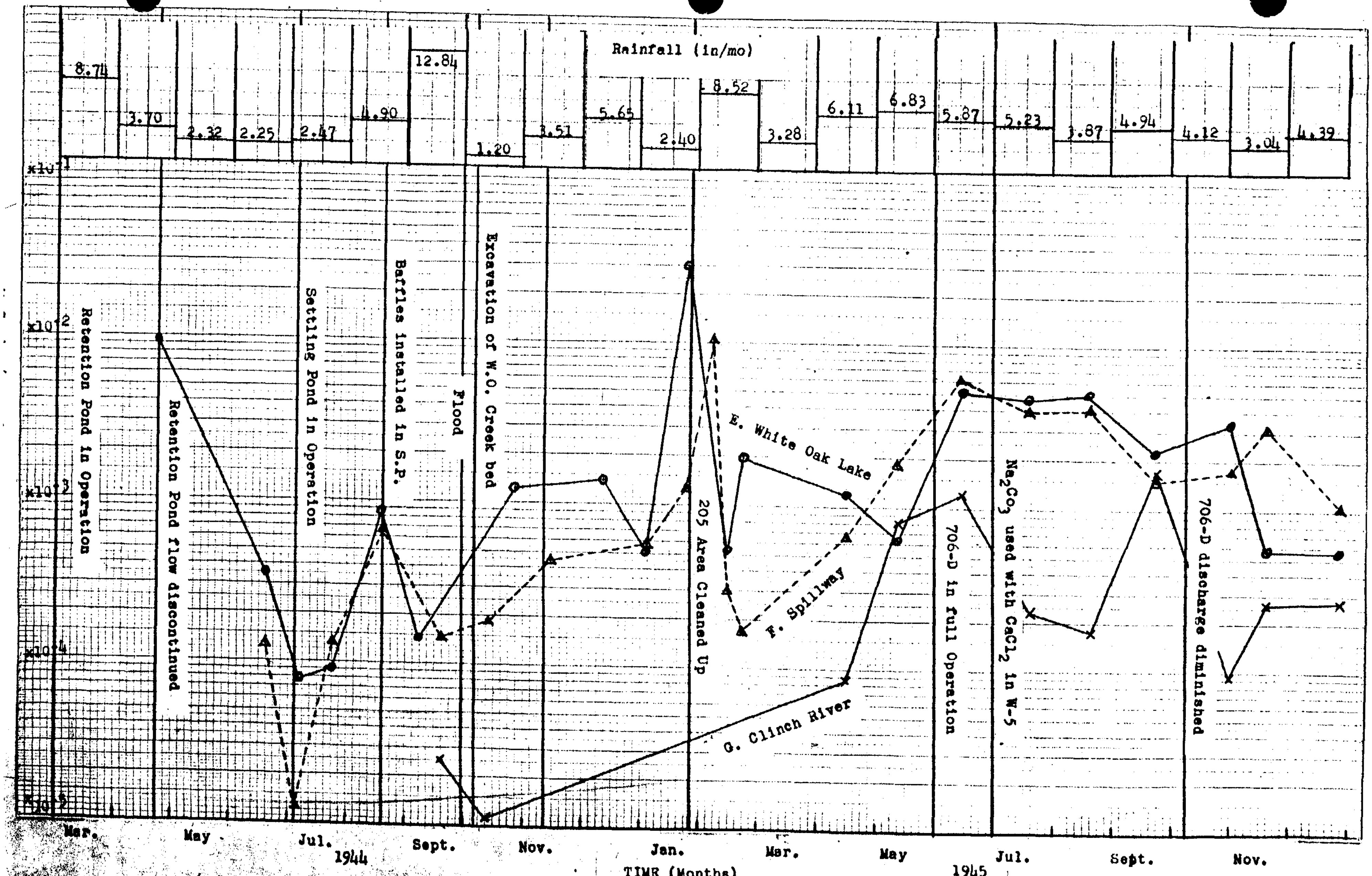

Fig. 4 SUMMARY OF WHITE OAK CREEK MUD DATA (W.O. Lake and below) 1944 & '45

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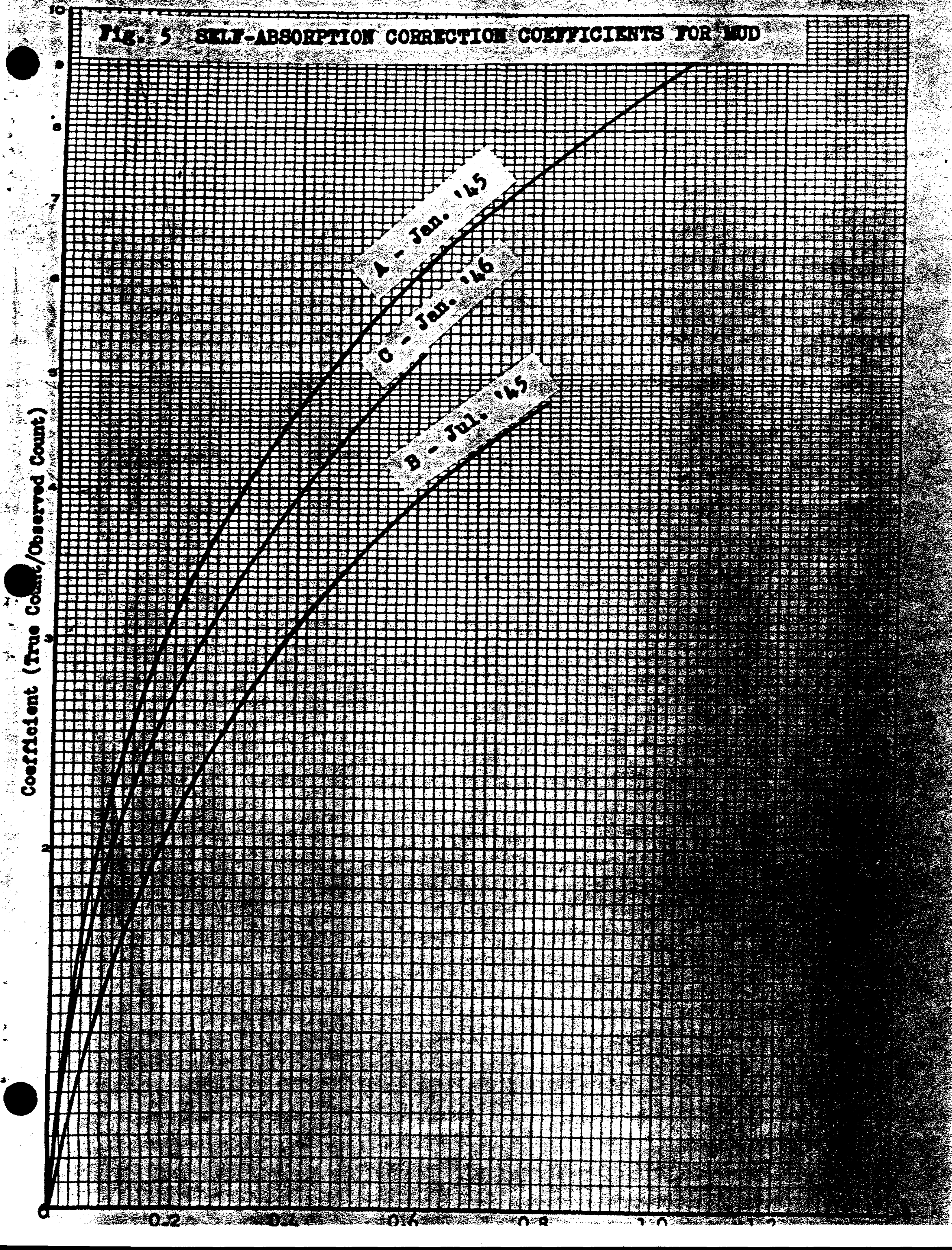
Fig. 5 SELF-ABSORPTION CORRECTION COEFFICIENTS FOR MUD
A - Jan. '45
C - Jan. '46
B - Jul. '45
Coefficient (True Count/Observed Count)
0
2
4
5
6
7
8
9
10
0.2
0.4
0.6
0.8
1.0
1.2

changes took place in operations. A monthly summary of rainfall has also been added to each chart.

Several phenomena are immediately evident from these charts. Mud activity after 7 weeks of discharge through the retention ponds had reached approximately 0.4 microcuries per gram at points as far down as the marsh section. Six weeks without discharge brought about a drop in the radioactivity of the mud in the upper portion of the drainage system by a factor of from 200 to 800, principally due to a scouring effect. A survey taken a few days after discharge was resumed, using the new settling pond, showed that the activity at the upper end of the creek had risen to about 10% of its high value on April 26, 1944. Effects of this discharge were not apparent at the lake, and below, until about a month after it was begun.

The mud activity rose somewhat throughout the drainage system during the summer. It was noted that algae, growing at the bottom of the settling pond were becoming dislodged by CO_2 bubble flotation, and were passing out of the pond through the outlet weirs carrying considerable quantities of radioactive material. Baffles, shielding the outlet weirs, were installed in August, 1944, and subsequent surveys showed mud activity downstream to have dropped by a factor of 2 to 8.

The next significant change in mud activity was produced by the flood of September 29. At this time the creek overflowed its banks and a considerable stream of water flowed through the settling pond. It is probable that a great amount of activity was entrained by this stream, but the enormous rate of dilution made it impossible to detect any significant increase in concentration of radioactive isotopes in the water. Much of this excessive contamination was deposited downstream in the clay of White Oak drainage system, as is indicated by a sharp rise in curves of Figures 3 and 4. The radioactive isotopes that were entrained by the mud did not cause a net increase in the mud activity of the Clinch River. White Oak Creek was deepened and widened during October and November, for a distance of a few hundred yards where it passed the settling pond, to enable it to accommodate future floods without jeopardizing the pond. In the course of the excavation, the small pond previously mentioned at point A was destroyed. Consequently, due to the flood and the excavation, the radioactivity upstream at point A decreased while values downstream increased as a result of the radioactive mud washed from upstream.

A sharp rise in activity appeared in the creek bed during January, 1945, when the Plutonium Separations Building was cleaned up after the cessation of separation operations. This radioactivity subsided in a short time, and values remained fairly constant at the December level until the barium separations began in volume in Buildings 706C and 706D. The radioactivity in the Clinch River mud at the mouth of White Oak Creek began rising early in 1945, and has followed somewhat erratically the fluctuations in the radioactivity of the mud in White Oak drainage system.

The 706C and 706D Buildings' chemical wastes are the result of different isotope separations from young slugs and behave differently in the cooling tanks, settling pond and drainage system from those resulting from the separation of plutonium. As a result, the precipitation of active constituents upon dilution occurs to a lesser degree, and about half or more of the curie content of the supernate that is jetted out of tank W-6 passes through the settling pond and into the White Oak Basin. As a result, when building 706D was put into full operation in June, 1945, enough contamination was carried through the settling pond to increase the mud activity by a factor of about 10 throughout the drainage basin. Also, the half distance increased to about 2400 feet under these conditions, indicating a decrease in the percentage decontamination per unit distance. It may be noted also that grouped readings of the radioactive mud at G, the junction of White Oak Creek with the Clinch River, increased in the course of one year by a factor of almost 100, reaching 1.7 x 10^{-3} μc/gm in September 1945.

The addition of $CaCl_2$ to tank W-5 was resumed in July. This caused a slight drop in creek mud contamination, reaching a reduction factor of 5 by the end of December.

4. Fission-Product Assay

During the months of March and April, 1945, an assay of fission-product contamination was made throughout the whole White Oak drainage system by L. H. Weeks. (4) The first problem was to make a careful survey of all the drainage area and to mark off a grid to aid in systematically locating sampling points. The assay was made by securing core samples of mud at these regular intervals, mixing the core samples to eliminate stratification, and determining the curie content of each sample by counting the beta-gamma activity in the usual manner with a mica window Geiger-Muller counter.

The samples were obtained by means of a piece of 2 inch i.d. iron pipe, turned to a taper at the end so that it could be pushed into the mud without disarranging the stratification. Preliminary tests were made on several cores to determine how deep a sample would have to be taken. It was found that, except for the old stream bed above the upper dyke and the channel in White Oak Lake, most of the activity was contained in the 2 to 6 inches of a core. When sampling, the bottom of each core was tested. If the radioactivity was not found to be zero, a deeper core was obtained at that point.

Two test samples were taken and counted from each thoroughly mixed core sample. If the two samples did not check, the core was mixed further and retested. The balance of the core was then dried and weighed, and the total curie content calculated. From this result, the number of micro-curies per square foot was calculated. In counting, the time for approximately

10,000 counts was used, except that the counting times were limited to ten minutes if the samples were not active enough to give 1000 c/min.

The above procedure was repeated in October 1946, by H. R. Craft and the results of these two surveys are given in Table I.

Table I

Total Radioactivity of White Oak Drainage System

Location	Approximate Distance Below Settling Pond (ft.)	April 1945		October 1946	
		Av. $\mu c/ft^2$	Total Activity (Curies)	Av. $\mu c/ft^2$	Total Activity (Curies)
Marsh	2,120	91.5	42.7	145.5	67.8
Intermediate Pond	3,250	87.6	4.6	-	-
W. O. Lake Mud Flats	6,900	22.0	14.9	9.6	6.5
W. O. Lake (south of trees across lake)	8,600	8.5	6.8	17.1	13.6
[illegible] below spillway	10,350	1.04	0.3	2.7	0.9
Probable Total Curies -			69.3		95

It was estimated by W. D. Smith [6] that about 5 curies/day were discharged into the Creek during the 52 days that the retention ponds were used, Morgan and Western [1] indicate that a total of about 520 curies were discharged from the beginning of operations of Clinton Laboratories until the time of the April 1945 general mud survey. Thus, about 13% of the activity discharged into White Oak drainage system during a year and a half of operation still remained in the mud in April 1945. During the seventeen months between this and the October 1946 survey, an estimated additional 890 curies were discharged into the system, making a total of 1410 Beta curies since the beginning of operation. Of this quantity about 7% was detected during the October 1946 survey.

It is never possible to make an accurate estimate of the curies one would expect to find in the White Oak Drainage system. However, a very rough estimate has been made on the basis of two assumptions; first, that all the radioactive isotopes that escaped from the Settling Pond were deposited on the clay of the basin, and second, that the precipitation in the pond was uniform and consistent with experimental results obtained by R. S. Apple [7], who ran experiments to determine what part of the original isotope content of W-6 supernate would precipitate on dilution with pond water. Using a distribution based on Apple's analysis of unprecipitated isotopes at 35:1 dilution, curie content on April 26, 1944, at the end of 52 days of discharge through the retention ponds was calculated by adding the results of integrating $\frac{dc_i}{dt} = K_i - \lambda_i c_i$ from $t = 0$ to $t = 52$, where K_i, λ_i and c_i are the average daily discharge, the decay constant, and the curie content, respectively, for each radioactive isotope. Discharges for the months subsequent to the opening of the Settling Pond were assumed to have the same fission-product distribution, and for simplicity of calculation the total monthly curie discharge was assumed to take place in the middle of the month. Then the curie content, C, present at the time of the April 1945 assay would be $\sum C_i = \sum C_{oi} e^{-\lambda_i t}$, when t is taken individually for the batch present on April 26, 1944, and that of each month since the Settling Pond has been in operation. These calculations gave 97.5 beta curies as the approximation of the amount which might be expected to be present at the time of the April 1945 assay.

Several discrepancies exist in the assumptions used as bases for the estimate. The retention ponds, as previously mentioned, lost considerable quantities of active precipitate at times of drainage; various amounts of radio-isotopes escaped into the Clinch River, as shown by Morgan and Western [1]; and the analysis of Apple [7] was probably valid only during the time that the plutonium separation operations were the primary source of wastes passing through the Settling Pond, i.e., through January 1945. From these considerations it appears that the figure of 97.5 curies is a low estimate of the discharged curies remaining in April 1945 after considering the [illegible] decay. Consequently, one might conclude that the White Oak basin is, at best, not over 70% efficient as a decontaminating agent for radioactive plant wastes.

5. Self-Absorption in Mud

A major potential source of error in evaluating mud activity is the self-absorption of the sample itself. This is a variable source of error, depending on the thickness of the dried sample as prepared for counting, and the type of mud. In magnitude, it may vary the resultant by a factor of 2 to 4 in the normal range of thicknesses of samples which is 0.15 to 0.3 gm/cm^2.

The self-absorption coefficient was determined by preparing a series of samples from the same specimen, ranging in thickness from about 0.025 gm/cm^2 to about 1 gm/cm^2. The values of c/min/gm were plotted as a function of gm/cm^2 on semi-log paper and the resulting curve extrapolated to zero thickness of sample. From this extrapolation, the self-absorption coefficient or the ratio of true count to actual count was estimated.

Figure 5 shows the results of several determinations made at different times. Curve A is the resultant of several random samples collected in January, 1945. Curve B is for samples collected in July, 1945, by A. T. Greenwood. It is obvious that self-absorption is considerably less at the latter date. The self-absorption depends chiefly on the energies of the radiation encountered, and to some extent upon the ratios of beta to gamma activity. In general, among beta emitters, a long-lived element has a softer radiation than one with a short life, and the difference in Curves A and B is consistent with the previously mentioned increase in short-lived constituents in the chemical wastes due to the barium separation operations in 1945. It frequently happens that there is selective adsorption of some of the radioisotopes in the mud. This property varies with the chemical properties of the water and mud, and with the plant life present in the mud. The thin window counters used are about 100 times more sensitive to betas than to gammas when thin samples are used, but a thick sample tends to accentuate the gamma counts. Curve C of Figure 5 represents a similar determination in January 1946.

It appears necessary to make a determination of the self-absorption coefficient periodically, especially whenever a change in operations brings about a change in the composition of the chemical wastes.

6. Chemical Analyses for Specific Activities

Chemical analyses were made occasionally to identify and evaluate the relative amounts of the various fission products present in the mud of the White Oak drainage system. These analyses give some indication of the effectiveness of the mud in the removal of specific activities from the water wastes.

The first samples analyzed were those taken on 4/26/44 by Overstreet and Jacobson.[2] The results are given in Table 2.

Table 2

Element	Sample D (marsh)	Sample E (Int. pond)	Sample F (lake)
Ba and Sr	.88%	.98%	1.42%
Zr	46.7	47.2	43.7
Cb	34.9	32.6	34.8
Ce	12.8	13.8	14.1
Y & Pr	4.71	5.46	5.96
Gross c/min/gm	41,200	6,840	1,220

Samples collected on 6/10/44 and analyzed by J. G. Hamilton gave the results shown in Table 3.

Table 3

Element	Sample 2 (S.P. outlet)	Sample 12 (marsh)	Sample 22 (lake)
Ba & Sr	.70%	.98%	1.4%
Zr	55	47	41
Cb	28	32.2	38
Ce	12	14	13
Y & Pr	4	5.5	5.6
Gross c/min/gm	42,500	8,490	1,540

An analysis by D. M. Black on samples collected 4/14/45 showed a change in specific activities. The results are given in Table 4.

Table 4

Element	Sample 2	Sample A	Sample 1B	Sample 4B
R.E.(mostly Ce)	62.5%	50.0%	73.0%	71.5%
Sr	6.3	7.4	1.3	.9
Zr	9.4	19.1	9.4	8.3
Cb	7.3	10.3	1.9	5.5
Ru	14.6	13.4	14.5	13.8
Gross c/min/gm	260	717	1,370	1,760

The ratio of the activity of strontium to the other specific activities is considerably higher in this test than in those summarized in Tables 2 and 3. However, considering the small value of the gross activity, the results of this test are less significant, both because of the larger probable error and the small absolute value of strontium present.

Table 5, representing results of analyses made by Black on 8/15/45 and J. E. Hudgens on 2/14/46, respectively, shows a significant rise in the specific activities of barium, strontium and cesium. These results show further the radical difference in the chemical wastes from the Barium Separations Plant in Building 706D from those of the plutonium separations carried out in Building 205 until February 1945.

Table 5

Element	Marsh Sample 8/15/45	Sample A (marsh) 2/14/46	Sample B (Int.P.) 2/14/46	Sample C (lake) 2/14/46
Ba	8.1%	19.4%	12.9%	
Ru	21.0	15.2	14.3	
Sr	10.1	15.9	23.1	36
R.E.	27.9	46.7	40.7	
Cb	.04			
Zr	1.1	0.9	.8	
Cs	31.7	10.6	11.1	
Gross c/m/g	2269	1287	753	

Table 6 represents estimated total quantities of barium, strontium and cesium on various dates. These values are calculated from percentages of these elements, as shown by chemical analyses, and total activities of the mud, as shown by standard survey methods on the respective dates.

Table 6

Date	Av. total μc/gm in mud of marsh	Probable μc/gm of (Ba, Sr & Cs) in marsh	% (Ba, Sr, & Cs)
4/26/44	3.7×10^{-1}	3.25×10^{-3}	.88
6/10/44	2.3×10^{-3}	2.26×10^{-5}	.98
4/14/45	3.4×10^{-2}	4.42×10^{-4}	1.3
8/15/45	4.8×10^{-2}	2.42×10^{-2}	50.4
2/14/46	1.28×10^{-2}	5.88×10^{-3}	45.9

[redacted]

Tellurium was not identified in any of these analyses, although Overstreet and Jacobson(2) reported that W-6 supernate contains 4.29% of Tellurium.

In an overall evaluation of the White Oak Creek basin as a part of the waste water decontamination process, one might conclude that it has served its purpose well. With the proposed changes of process, and consequent changes in the nature of the chemical wastes, further changes in the decontamination of wastes may be required in order to keep the White Oak Lake effluent within safe limits. In general, it seems that a slowly flowing drainage system like White Oak Creek and lake with a clay bottom is an economical and rather efficient means of removing fission products from plant waste water.

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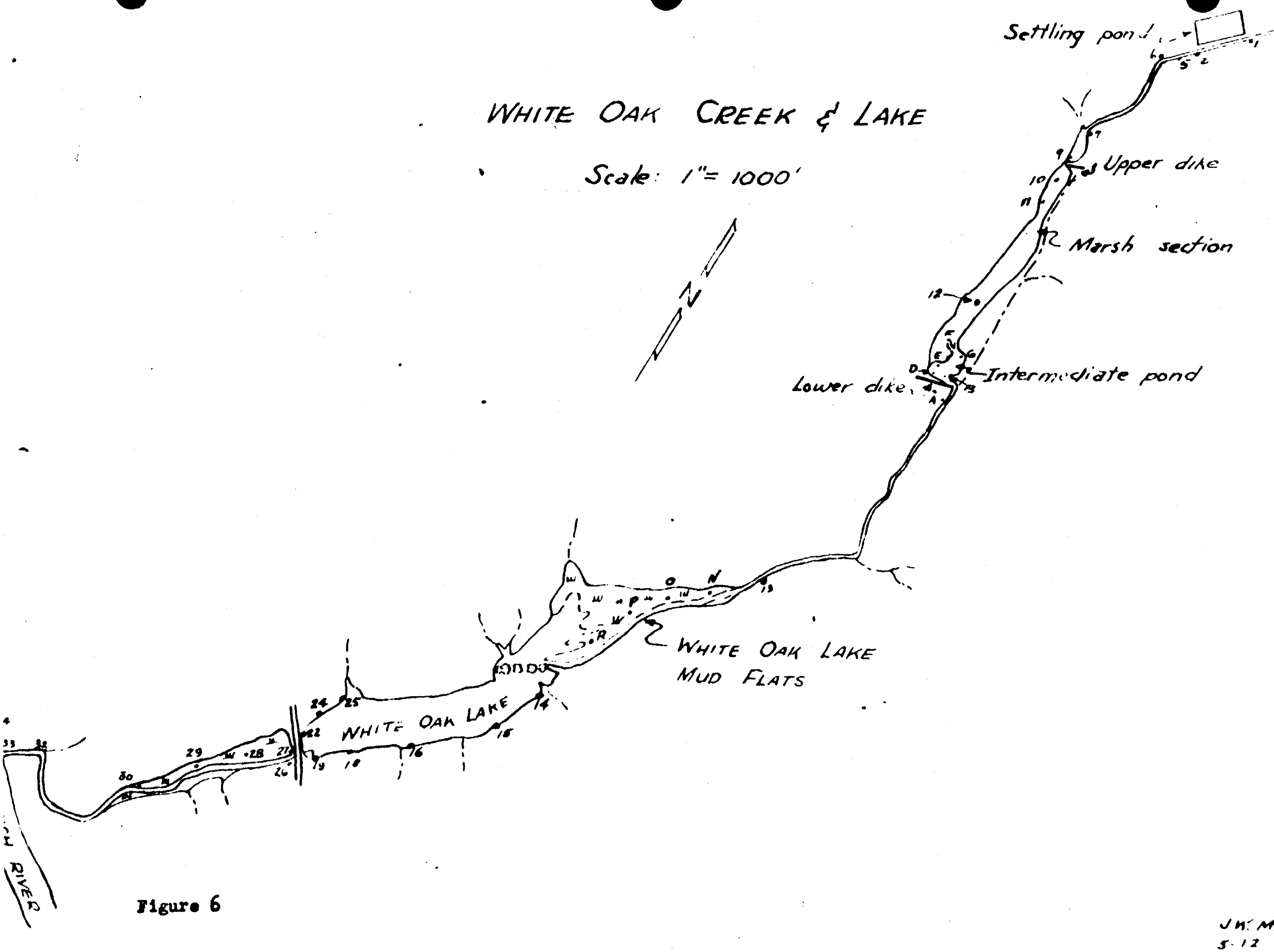
WHITE OAK CREEK & LAKE
Scale: 1" = 1000'
N
Settling pond
Upper dike
Marsh section
Intermediate pond
Lower dike
WHITE OAK LAKE
MUD FLATS
WHITE OAK LAKE
RIVER
J.W.M.
5-12-45

Figure 6

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